

## LIQUID CURABLE POLYORGANOSILOXANE COMPOSITIONS

### REFERENCE TO RELATED APPLICATIONS

This application is a continuation-in-part of application Ser. No. 592,481, filed Mar. 23, 1984, now abandoned. This application was filed under the provisions of 37 CFR 1.62.

### BACKGROUND OF THE INVENTION

#### 1. Field of the Invention

This invention relates to liquid polyorganosiloxane compositions which are curable by means of a hydrosilation reaction to yield tough elastomeric products.

#### 2. Description of the Prior Art

Liquid compositions prepared by combining a vinyl-containing polydiorganosiloxane, an organohydrogen-siloxane curing agent, a platinum-containing catalyst and, optionally, a filler are known in the art. These liquid compositions can be cured under relatively mild conditions to yield elastomeric or resinous products, depending upon the types and relative concentrations of reactants present in the initial curable composition. These curable compositions are useful in a variety of applications, including encapsulation of delicate electrical and electronic components, coating of various substrates, as pressure sensitive adhesives, and the formation of shaped articles by injection molding.

Reinforcing fillers such as fume silica have been used to improve the physical properties such as tensile strength, tear strength and modulus of cured polyorganosiloxane compositions.

The use of resinous siloxane copolymers as a replacement for reinforcing silica fillers to improve the physical properties of cured articles while retaining the transparency of an unfilled composition is taught in the prior art. Specifically, U.S. Pat. No. 3,284,406 to Nelson, which issued on Nov. 8, 1966, and U.S. Pat. No. 3,436,366 to Modic, which issued on Apr. 1, 1969, disclose using resinous copolymers containing  $R_3SiO_2$ ,  $R_2ViSiO_2$  and  $SiO_{4/2}$  units for this purpose. In the foregoing formulae, R represents a monovalent hydrocarbon radical free of ethylenic unsaturation and Vi represents a vinyl radical.

The cured materials disclosed in the aforementioned Nelson and Modic patents typically exhibit hardness values of from 35 to 80 on the Shore A durometer scale, which is indicative of relatively highly crosslinked materials. Modic teaches inclusion of a finely divided non-reinforcing filler when transparency of the cured article is not a requirement. The curing agents taught by Modic are liquid polyorganohydrogensiloxanes containing at least two silicon-bonded hydrogen atoms per molecule, while the curing agents disclosed in the aforementioned Nelson patent contain at least three silicon-bonded hydrogen atoms per molecule.

A means for increasing the tear strength of cured polyorganosiloxanes prepared from liquid compositions similar to those disclosed in the aforementioned Modic patent while maintaining the viscosity of the curable composition within the range from 10 to 500 Pa.s is taught in U.S. Pat. No. 4,340,709 to Jeram and Smith, which issued on July 20, 1982. In accordance with the disclosure of this patent, a liquid polydiorganosiloxane containing from 0.14 to 2.0 mole percent of vinyl radicals is cured using the combination of a crosslinking agent and a "coupler." The coupler is a linear polyor-

ganosiloxane containing only two silicon-bonded hydrogen atoms, one at each of the two terminal positions of each molecule. The crosslinking agents are defined as "hydride resins having only terminal hydrogen atoms or a linear hydride polysiloxane" containing hydrogen atoms only in the internal portion of each molecule on nonterminal silicon atoms.

Jeram and Smith limit the concentration of difunctional coupler to the range of 4 to 15 parts by weight per 100 parts by weight of vinyl-containing polydiorganosiloxane. The coupler is further defined as providing a molar concentration of silicon-bonded hydrogen atoms that is from 0.67 to 1.3 times the molar concentration of vinyl radicals present in the curable composition. The cured compositions exemplified by Jeram and Smith exhibit hardness values of from 21 to 43 on the shore A durometer scale after curing for one hour at 100° C. Hardness values in this range are desirable for protective coatings, encapsulating materials and certain types of molded articles, however the cured products disclosed by Jeram and Smith would not be useful for fabricating cured gel type materials which are required to be compressible under relatively low pressure.

The concept of using a difunctional curing agent in combination with a tri- or higher functional curing agent, both of which contain silicon-bonded hydrogen atoms, to improve the tensile properties of cured polyorganosiloxane elastomers is taught in U.S. Pat. No. 3,697,473, which issued to Polmanteer et al. on Oct. 10, 1972. The elastomers exemplified in this reference exhibit durometer values of from 11 to 38, measured on the Shore A scale. At least 10% of the available silicon-bonded hydrogen atoms are present in each of the two types of curing agents.

The tensile properties and hardness values reported in the aforementioned Nelson, Modic, Jeram et al. and Polmanteer et al. patents indicate that the cured articles exemplified in this prior art do not exhibit the unique combination of properties required for some applications of polyorganosiloxanes, for example, elastomeric optical waveguides.

Pressure sensitive optical waveguides formed from elastomeric polyorganosiloxanes are disclosed in U.S. Pat. No. 4,050,027, issued to Pfister and Maher on Sept. 20, 1977, and U.S. Pat. No. 4,128,299, issued to Maher on Dec. 5, 1978.

Pressure sensitive optical waveguides require cured, noncellular articles in the form of blocks or sheets that are transparent, exhibit minimal light attenuation, and are deformable by an electrostatic or mechanical force applied against at least one surface of the article. Polyorganosiloxanes suitable for this application typically exhibit durometer values too low to be measured using the Shore A scale. In addition, the waveguide must be sufficiently resilient to resume its original shape almost immediately following release of the pressure which deformed it, and sufficiently tough to resist being punctured and/or torn when this pressure is repeatedly applied over only a fraction of the total area of one surface on the waveguide. If the surface of the waveguide on which deformation occurs is overlaid with a film or membrane such as the cladding of an optical fiber or optical waveguide, the adhesion along this interface must be sufficient to prevent separation between the two components and resultant void formation. The foregoing combination of properties considered desir-